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1. Your reference — HMJ03612GB

2. Patent application number *(The Patent Office will fill in this part)* — 0224447.3

3. Full name, address and postcode of the or of each applicant *(underline all surnames)*

   Ole-Bendt RASMUSSEN
   Sagenstrasse 12
   CH 6318 Walchwil
   SWITZERLAND

   Patents ADP number *(if you know it)* — 7916943001

   If the applicant is a corporate body, give the country/state of its incorporation

4. Title of the invention — Crosslaminate of Oriented Films, Method of Manufacturing same, and Coextrusion Die suitable in the Process

5. Name of your agent *(if you have one)* — Gill Jennings & Every

   "Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)*

   Broadgate House
   7 Eldon Street
   London
   EC2M 7LH

   Patents ADP number *(if you know it)* — 745002

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Country	Priority application number *(if you know it)*	Date of filing *(day / month / year)*

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Claim *(s)*	8
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Request for substantive examination *(Patents Form 10/77)*	NO
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11. For the applicant
Gill Jennings & Every

I/We request the grant of a patent on the basis of this application.

Signature

Date
21 October 2002

12. Name and daytime telephone number of person to contact in the United Kingdom

H M M JONES
020 7377 1377

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Crosslaminate of Oriented Films, Method of Manufacturing same, and Coextrusion Die suitable in the Process.

The present invention represents a widening of the scope of the invention claimed in British Patent Application No. 0205021.9 filed on March 4th, 2002. Extracts from that specification are included herein, after the example of the present i This copending application deals with a crosslaminate of oriented films in which the bonding exhibits a strongbond/weakbond or a strongbond/nobond pattern. The purpose is to combine a high tear propagation resistance with a high resistance to delamination, a combination of properties which otherwise is difficult to achieve for thin crosslaminates, e.g. crosslaminates of a gauge lower than generally about 100 gr. per sq.m. In the copending application, the locations of strong bonding are formed by coextruding an array of thin strands on the main layer of two films (A and B) used for the crosslamination, select the polymer or polymers for these strands suitably for the bonding, and arrange the films for lamination so that strand direction and main direction of orientation in A criss-crosses with strand direction and main direction of orientation in B, while the strands on A face the strands on B. Under use of heat A and B bond together at least in the spots where the strands on A intersect with the strands on B. Preferably the criss-crossing arrangement is achieved under use of spiral cutting of a tubular, coextruded film and/or under use of a relative rotation between the exit of an annular coextrusion die and the means to take up the tubular film after the extrusion

However, the extrusion can also be carried out under use of a flat coextrusion die, but the lamination then must take place as a socalled "cross-webbing" process, which is more complicated.

Between the mainlayer and the strands in each of the films A and B there is preferably coextruded a continous layer, referred to as the "second surface layer", of a composition which is selected to produce some bonding, but a weak bonding, in the areas which are devoid of strand material.

The above mentioned copending patent application the text of which is incorporated below also mentions that the disclosed annular coextrusion die which is preferred for coextruding of stripes, can be "applied in the production of polymer film other than crosslaminates e.g. film with a decorative pattern of coloured stripes". In this connection it should be clear that there can be ~~estetical~~ aesthetic values in colouring the stripes in the criss-crossing bonding pattern, provided the criss-crossing pattern can be seen at least from one side of the laminate (remembering that the criss-crossing stripes are not on the surfaces of the laminate, but are inside it). Upon further consideration the inventor has realised that this ~~estetical~~ aesthetic effect may have equal or perhaps even bigger commercial importance than the technical effect (the combination of high tear propagation resistance and high resistance of delamination) and that consequently the scope of the claimed invention should be widened to cover a crosslaminate with so produced cross-crossing coloured strands inside, also in cases when

strongbond/weakbond or strongbond/nobond pattern. In this connection it is the experience of the inventor that the raw material saving which can be achieved by use of crosslaminated film, often from a commercial point of view is offset by negative subjective judgements. As an example agricultural tarpaulin (e.g. for protection of crops) made from a 70 gm pr.sq.m. crosslaminate of oriented polyethylene films would be a fully adequate substitute of a 100 gsm tarpaulin made from extrusioncoated woven tape, if only objective criteria were applied.

However, in actual fact the average customer to agricultural tarpaulins makes his choise to a great extent on the basis of the "handle" and the appearance, and will reject the 70 gsm tarpaulin due to its flimsiness and its look as a "simple plastic film". The problem of flimsiness is reduced by application of the inventor's earlier invention described in ~~EP-A-0622412 6~~ WO-A-9314928, which briefly is explained below in connection with one of the special embodiments of the present invention, while the problem of "look as a simple plastic film" still has been outstanding. However it is believed that the pattern of criss-crossing strands will convey the message that this is a crrosslaminate and therefore of special strength. In this connection it can be seen that the pattern clearly is not printed on the surface of the laminate but is inside it. The slightly blurred delineation of the pattern, which is an inherent result of the coextrusion technique, shows that the lines come from

more plies. All in all the pattern signalises that this is a crosslaminate and therefore can be expected to be strong. Furthermore the pattern will of course be unaffected of abrasion, while a pattern printed on the laminate will be very susceptible ~~suspectible~~ to such actions.

A very big proportion of the crosslaminated film which is manufactured worldwide, has been stretched transversely by passage between one or more sets of intermeshing grooved rollers (see e.g. the above mentioned WO-A-9314928 ~~EP-A-0624126~~ which makes record of the then existing technique in connection with crosslaminated films). This transverse stretching process will always more or less give the crosslaminate a striated appearance due to surface corrugations with corresponding thickness variations in the laminate. A fine pattern of variations even as small as +/-5% becomes very visible due to reflections. The inventor has now surprisingly found that the combination of this striated pattern of coloured strands inside the laminate produces a pronounced 3-dimensional effect. Besides being an interesting effect which can attract curiosity it also make the observer feel that the laminate is much thicker than it really is, and thereby it works against the negative subjective judgement that this is "simple plastic film". This special effect is further dealt with bellow.

In its present widened scope, the invention concerns a crosslaminate comprising mutually bonded polymer films of which at least two neighbour films A and

unbalanced biaxially oriented, whereby the main direction c orientation in A crosses the main direction of orientation in B and each contains a layer consisting of a polymer material selected for high tensile strength (hereinafter th main layer) and on each mainlayer on its side which faces the the neighbour film A or B at least a first surface layer. Said first surface layer on each of the films A and is a discontinous layer consisting of an array of coextrude thin strands consisting of a material which is selected to modify the properties in the surface of the respective film This modification concerns either the optical appearance of the laminate or the bonding between A and B.

Accordingly, in the method of manufacturing the crosslaminate of the invention, which laminate comprises th above mentioned films A and B (but there can also be furthe films in the laminate), A and B are each coextruded in a flat or circular coextrusion die, and each comprises a main layer of a polymer material which is selected for high tensile strength and the above mentioned first surface laye made from a different material. A and B are each supplied with a uniaxial or unbalanced biaxial molecular orientation at any stage after the joining of the different materials in the coextrusion die and before the lamination. Prior to the lamination A and B are arranged in such a way that the main direction of orientation in A will cross the main direction of orientation in B, and during the lamination the bonding between A and B is established at

the method is that in the coextrusion each of the said first surface layers is made a discontinuous layer (discontinuous transversely to the direction of extrusion) consisting of an array of strands, and in the lamination the array of strands on A are arranged to cross the array of strands on B. The method is further characterised in that the material from which the strands are extruded is selected to modify the properties in the surface of the respective film. This modification concerns either the optical appearance of the laminate or the bonding between A and B.

The coextrusion of one or both films A and B is preferably carried out by means of a circular coextrusion die, to form and draw-down a tubular film. Hereunder the draw-down is adpated to produce a significant uniaxial or unbalanced biaxial meltorientation with the main direction of orientation and the direction of the array of strands along the longitudinal direction of the film. Alternatively, the orientation and the direction of the array can be made to extend helically along the tubular film by means of a relative rotation between the exit of the die and the means to take up the film after the extrusion. Subsequently the film is out open under an angle to the main direction of orientation and to the direction of the array. The distance from middle to middle of neighbour strands at the exit from the extruder should normally be at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm, and the circumference of the tube at this ... at least 20 cm.

Following the bringing-together of the films in a sandwich arrangement for lamination, before or after the bonding of said sandwich arrangement to a laminate by heat, the films may become further oriented by stretching in the longitudinal and/or in the transverse direction. Such steps are not new in themselves - see e.g. the above mentioned EP-
WO-A-9314928
~~A-0624126~~ - but can provide particular advantages in connection with the present invention.

In the lamination process the strands in A can be directly sealed to the strands in B, but alternatively the lamination process can be extrusion lamination whereby the bonding is established by means of a separately extruded layer.

In order to avoid a reduction of the tensile properties of each of the films A and B, the strands should normally occupy at the highest 30%, preferably at the highest 20% and still more preferably no more than 10% of the respective film, in the thickness of the film and/or across the area of the film.

The distance from middle to middle of neighbour strands in each array should normally be between 2 mm and 80 mm, preferably no higher than 40 mm, and more preferably no higher than 20 mm.

Normally the array of strands on each of the films A and B should occupy no more than 15%, preferably at the highest 10%, and more preferrably at the highest 5% of the volume of the respective film A or B.

In addition to the strandformed "first surface layer" there

coextruded a second surface layer on the mainlayer of at least one of the neighbour films A and B on the side of the mainlayer which faces the other one of the said neighbour films. This second surface layer should be continuous and can either either be located between the mainlayer and the first surface layer or on top of the first surface layer. It is preferably selected for control of the bonding between A and B.

On one or each of the outer films of the laminate, there should normally be coextruded a surface layer which also will be surface layer of the laminate, and which is adapted to enhance heatsealing of the laminate and/or increase its frictional properties.

Each of the said two films A and B will normally mainly consist of polyethylene or polypropylene.
Except for the situation that there is coextruded a second (continuous) surface layer on top of the strandformed first surface layer, it will always for the sake of easy or improved lamination, be advantageous to select the composition of the strand material so that the strands, generally speaking, melt at a temperature at which the mainlayer still is mainly solid. Thus, more exactly expressed, the average melting point of the polymers which constitute the strandformed first surface layer, should normally be at least about 10°C, preferably at least about 15°C, and more preferably at least about 20°C lower than the

main layer.

In this connection, the "average" should of course be understood as an average with different weights on the constituents. Thus if the strands consist of 20% of polymer X having melting point 125°C and 80% of polymer Y having melting point 90°C, its average melting point will be

$125 \times 20\% + 90 \times 80\% = 97°C$.

In one aspect of the invention the purpose as mentioned is to provide a strongbond/weakbond or strongbond/nobond system of lamination, and thereby a suitable combination of tear-propagation-resistance and resistance to delamination. Several embodiments of this aspect are dealt with in the copending British Patent Application and shall not be repeated here. However, a further advantageous embodiment should be mentioned. In this embodiment the weak bonding, which here perhaps better can be called a blocking, is established by means of an polymer addition in the 2nd surface layer of a polymer type as used as addition in cling film. Such addition is a sticky polymer, e.g. polyisobutylene, normally of a relatively low molecular weight, which shows a tendency to migrate to the surface. As an example of another suitable polymer for this purpose atactic polypropyelene can be mentioned. The contents of this addition should of course not be so high that the sealing between this second surface layer and the strandformed first surface layer is ruined.

Turning now to the other aspect which concerns the optical appearance, and in which it is essential that the strands ... are coloured and visible from one or

both sides of the laminate, it has already been mentioned that there appears an interesting, advantageous and surprising 3-dimensional optical effect when the surface of the laminate through which the strands are observed is embossed in a striated pattern. Normally the embossment needs not be made specially for this purpose, but can be a result of the transverse stretching between grooved rollers which today is widely used in the manufacture of crosslaminated film, see e.g. WO-A-9314928 & EP-A-0624126 (Rasmussen).
Especially it is the aim to make a crosslaminate, which has a general thickness of at the highest about 0,3 mm, appear as if the strands were at least about 0,5 mm distant from the striations. The wording "general thickness" is used because the striated embossment causes gauge variations. The "general thickness" within - say each cm extending perpendicular to the striations - should be understood as the average thickness within this span.
This embodiment of the invention is, more specified, defined as follows:

a) the laminate has an general thickness of at the highest about 0,3 mm,

b) A forms one surface of the laminate,

c) the laminate surface at least on the A-side exhibits a visible pattern of striations along one direction constitued by surface corrugations with corresponding thickness variations in A, the divisions in said pattern being at the highest about 3 mm,

d) the thin strands are coloured, and the rest of the film

A is sufficiently transparent to show the coloured strands when the laminate is observed from the A-side. ~~whereby~~ By this combination of features the depth of the corrugations is sufficient to make the strands appear as being at least about 0,5 mm distant from the striations.

Tentatively the 3-dimensional effect is explained partly as a psychological illusion, and partly by the fact that the stripeformed embossment form an alternating arrangement of structures which very approximately can be described as "cylindrical collection lenses" and "cylindrical spreading lenses". However, this must not be understood too strictly, since the "lenses" may more have the character of prisms, or there may be an alternation between lenselike segments and flat segments. The embossment may show a generally constant division, or may be more accidential, as when the embossments from several pairs of grooved rollers interfere with each other.

Whatever the explanation of the 3-dimensional effect may be (the explanation made above is tentative, as mentioned) the inventor has found this very significant as it also will appear from the example.

A particular strong optical or psychological effect of the striations can be achieved when the laminate, viewed in a cross-section perpendicular to the striations,

exhibits a generally regular arrangement of ribs which are thicker than the average thickness of the laminate and have a generally concave and a generally convex surface, so that ... bending of the rib transverse of its

longitudinal direction, and further so that the material in or adjacent to the boundaries of the ribs in the tensionless state of the material are bent in the opposite direction to the rib to give the material between the two adjacent ribs a generally straightened-out shape. This in itself is not new, but it is a main aspect in the above mentioned WO-A-9314928 (Rasmussen). In that patent the purpose is to give the laminate essentially improved stiffness in one direction, but in connection with the present invention it also provides a particular optical effect.

Particular aesthetic effects can also be achieved when the colour of the strands is formed by a pigment which supplies a metallic lustre or a iridescent effect. Masterbatches for such colours are usually very expensive, but when the application is restricted to the strands this is rather unimportant, and still the effect obtained in this way can be more estatic than the effect of an all over colouring of a film with such pigments.

According to a further aspect of the invention there is provided a new method of manufacturing a crosslaminate comprising mutually bonded polymer films of which at least two neighbour films A and B each are formed by coextruding in a flat or circular die a main layer of a polymer material which is selected for high tensile strength and a first surface layer from a polymer material, and in which A and B each is supplied with a uniaxial or unbalanced biaxial molecular orientation at any stage after the joining of the different materials in the coextrusion die and before the lamination, and prior to the lamination A and B are arranged in such a way that the main direction of orientation in A will cross the main direction of orientation in B, and during the lamination the bonding between A and B is established at least in part through heat, characterised in that in the coextrusion each of the said first surface layers is made

consists of an array of strands, and in the lamination the array of strands on A are arranged to cross the array of strands on B, and characterised in that the A film is provided with surface corrugations with thickness variations after its coextrusion, the divisions between the corrugations being no more than 3mm.

Preferably the surface corrugations are provided by transverse stretching of the laminate by intermeshing grooved rollers as described in WO-A-93/14928, so that the entire laminate has surface corrugations and these have the ribs described above. Preferably in this aspect the material from which the strands are extruded is coloured and the material from which film A is formed sufficiently transparent to allow the strands to be visible from the A side of the laminate.

The invention shall now be further described with reference to the drawings, of which:

Fig. 1 is a view approximately on a true scale of two coextruded, oriented, helical cut and crosslaminated films (A and B). It illustrates the aspect of the invention which concerns the optical appearance providing a 3-dimensional effect under use of coextruded strands (101) and (102) whereby a striated embossment (103) provides a 3-dimensional effect (The aspect which concerns bonding properties is illustrated in the above mentioned copending British Patent Application).

Fig. 2 shows a section through a-a in fig. 1. This section is perpendicular to the striations. The thickness of each layer is here shown in about ~~200~~ 400 times magnification, while the dimensions parallel to the surfaces of the laminate are shown in about ~~10~~ 20 times magnification.

Fig. 2a is a microphoto of the cross-section of the crosslaminate made as described in the example.

Fig. 3 is a schematic perspective drawing of a coextrusion line for manufacture of tubular film suitable, after helical cutting, for making the crosslaminate shown in figs. 1, 2 and 3. The flow of polymer material for the strands comes from a very small extruder (4), and flows of three other polymer materials (for main layer, continous "2nd surface layer" and layer for surface of the laminate) come from the bigger extruders (5), (6) and (7). The last mentioned three

materials are fed into the distribution part (8) of the coextrusion die and are here each formed into a circular flow. Joining of these flows takes place at the exit from (8) while they enter the bodily separate exit part (9). The flow from the small extruder (4) is fed directly into exit part (9) where, starting from the circumference, it becomes evenly distributed in a labyrinthine channel system as shown in fig. 5 and applied through a circular array of internal orifices on the outside of the tubular joint flow of the other components. (The concept of a "labyrinthine channel system" is explained in the above mentioned copending British Patent Application, and the name comes from US Patent 4,403,934).

Fig. 4 is an axial section, shown on about half scale, of the bodily separate exit part (9), which consists of the sub-parts (9a), (9b), (9c), (9d) and (9e). The section goes through the line x-x in fig. ~~4~~ 5. (10) are channels in the labyrinthine system.

Fig. 4a is a detail from fig. 4 showing the last branch in the labyrinthine channel system and one of the 64 internal orifices (11) through which the flows of strand-forming material join the tubular flow (12) of the three other coextruded materials. This detail is drawn on a scale about six times true scale.

Fig. 5 is a perspective view of sub-part (9a), showing the lower half part of the labyrinthine channel system (10) by

which one flow of the strand-forming material from extruder (4), fed through the inlet (13) stepwise is divided into 64 equal part-flows through the channels (10), each extruded separately through an internal orifice (11). The upper half part of the labyrinthine channel system, which is in sub-part (9b) is exactly symmetrical with that in fig. 5.

In fig. 1 the machine direction of the crosslaminated web is shown with the arrow (1). The main direction of orientation, which here is considered an unbalanced biaxial orientation, is shown by the arrow (2) for one film, and the arrow (3) for the other film. They are each shown under an angle of about ~~60°~~ 30° to the machine direction (1). ~~This~~ About 60° is what the inventor generally has found best for tarpaulins and cover sheet, while angles near 30° generally have been found best for crosslaminates used to make bags. The angle 45° was only found preferable in a few cases.

For each film the main direction of orientation is shown almost but not quite parallel with the array of strands (101) in film A and (102) in film B. These strands are in fig. 1 shown by thick interrupted lines. If the tubular film, when leaving the extrusion die, has been drawn-down straight, and if it has not been stretched after the helical cutting, the main direction of orientation will be exactly parallel with the array, but if the tubular film has been screwed during the ~~drawn~~ drawing-down, to produce a helical melt-orientation, or if it has been uniaxially or biaxially oriented after the helical cutting, as in US 4,039,364

quite parallel with the array.

Note that in fig. 1 the lines with reference number (103) are not hatching, but represent the striations formed by embossment, which appear in cross-section from fig. 2. This embossment is produced by stretching between grooved rollers, see the example. Due to this striation, the coloured strands (101) and (102) appear to be pronouncedly distant from the striations, even when the real distance is about equal to or even smaller than the resolution of the eye (which is about 0,1 mm). As already mentioned it is believed that this illusion is connected with the fact that the surface on the laminate in an alternating arrangement i concave and convex, or in other words, the film A, which here is considered transparent, consists of many fine "cylindrical collecting lenses" alternating with fine "cylindrical spreading lenses". The film B may likewise be transparent, but may also be deeply coloured to form an ~~estetic~~ aesthetic background for the pattern of coloured striations.

Fig. 2 further shows "2nd surface layers" (104) and (105) for control of bonding strength, and laminate surface layer: (106) and (107), made of polymer materials which are selected to improve the heatseal properties and/or the frictional properties of the laminate. In case the invention should be used not only for achievement of the optical effects, but also to establish a "strongbond/weakbond" pattern - this is normally the case -

the "2nd surface layers" should be between the respective
strandlayer (101) or (102) and the respective main layer
(108) or (109), as it is shown here, and as it further is
explained in the copending British Patent Application.
However, if there is not aimed at such "strongbond/weakbond"
effect, each strandlayer (101) or (102) can be coextruded
between the mainlayer (108) or (109) and the bonding
controlling "2nd surface layer" 104) or (105).
The cross-section shown in fig. 2 is drawn on basis of the
microphoto shown as fig. ~~4~~ 2a. The latter is explained in the
example. As already mentioned it is believed that the 3-
dimensional effect, which makes the coloured strands appear
much more distant from the striations than they really are,
is a result, at least in part, of the alternating
arrangement of the "cylindrical collecting lenses" and
"cylindrical spreading lenses" which constitute the
striations.

Figs. 3, 4, 4a and 5 have already been sufficiently
described for a principal understanding, but the following
should be added:
(14) shows different rows of bores for bolts or screws to
keep the parts strongly together. (15) in fig. 4 shows a
shallow channel for drainage, as usual in die construction.
In fact there should be a system of channels for drainage
occuping most of the area between sub-parts (9a) and (9b),
but for the sake of clarity only this channel is shown. With
reference to fig. 4a the downstream side of the internal
orifices (11) is given a shape which prevents damage of the

axial flow, a damage which can occur if there had been a sharp edge on this side of the orifices.

It has been emphasised that there is an exit part (9) bodily separate from the distribution part (8). As shown (9) will normally consist of several sub-parts. However, the shown sub-parts (9c) and (9e) can be constructed as one part. The centering of sub-part (9d) is made adjustable in order to compensate for thickness variations in the extruded tubular film (15).
The distribution of the flow from extruder (4) here is shown as a labyrinthine system, which is considered preferable, but it can also be other known circumferentially fed circular distributions systems.

~~In fig. 5 each~~ of the final 64 branches of the labyrinthine system ends in an internal orifice which extrudes directly into the axial tubular stream. However it is not practical to divide into more than 64 branches, and if a bigger number of strands is wanted, each final branch may open into a common ring-formed channel close to the channel (12) for the axial flow. From this ring-formed channel there can be the desired big number of openings into channel (12).

There may also be two (or more) labyrinthine systems of the described kind, one following after the other, and each ending in a circular row of internal orifices like (11), optionally with the above mentioned ring formed channel inserted between the final branches and the internal orifices. Preferably each such labyrinthine system should be fed from a separate small extruder. There may be used differently coloured polymer materials for the different small extruders. The internal orifices, which terminate the different labyrinthine channel systems, should be mutually displaced to avoid that the different sets of coextruded strands will cover each other.

aThus with reference to Figure 4, there can be inserted a further diering between rings (9a) and (9b), the surfaces of this inserted ring being shaped so that, together with (9a) it forms one labyrinthine system, and together with (9b) it forms another labyrinthine system.

Example of the present invention

The procedure is the same as in the above mentioned WO-A-9314928 ~~**EP-A-0624126**~~**, (Rasmussen) example 1 except for the following:**

The coextrusion line is constructed as shown in figs. 3, 4,

4a and 5, and there is coextruded strands consisting of a metallocene (catalysed) copolymer of ethylene and octene having melting range 50-60^{0}C and melt flow index 1.0. It contains 9% of a masterbatch for silver colour. This is about 3 times as much as normally used if the entire laminate should be coloured.

The layer which in said European patent is called "lamination layer" and here "2nd surface layer" is a blend of 90% LLDPE and 10% of the same low melting copolymer. The LLDPE has density 0,92 g per ml and melt flow index 1.0. Th "main layer" and the "heatseal layer" (for heatsealing of the final crosslaminate) are the same as in the said exampl 1.

The "main layer" forms 75%, the "heatseal layer" 15%, the "2nd lamination layer" 8%, and the strands 2% of the film. Only the strands are coloured.
The angle of cutting is 57^{0}.
The temperature for lamination, stretching processes, and final heat treatment are also different, namely:
For preheating: 60^{0}C
For the transverse stretching between the special grooved rollers described in the said European patent and the first longitudinal stretching processes: 50^{0}C.
For the following transverse stretching process: 35^{0}C. (Temperature of the grooved rollers).
For the heat treatment which effects the final bonding:

There is not in the above mentioned example 1 used a jet ai to cool the films during this stretching.

Like in the said example 1 the gauge of the final cross-laminate (for identification below called I) is about $70gm^{-2}$. This has the bonding pattern shown in fig. 1.

A similar crosslaminate (called II) but without the strands is manufactured for comparison.

Furthermore there is manufactured a third laminate (called III) similarly to I), but without the strands and with 15% instead of the 10% metallocene copolymer in the "2nd surface layer".

(I) exhibits the highest tear propagation resistance, (II) almost the same, and (III) a considerably lower tear-propagation resistance under shock-tearing, which is unacceptable for tarpaulins. This property is evaluated by "hand-tearing" at a meassured velocity between 5-7 ms^{-1} by a team of people used to such testing and knowing the requirements of customers. To the knowledge of the inventor there does not exist any standardised test for tear-propagation-resistance which comes close to the practical conditions of tearing.

(I), (II) and (III) are also cut into 20 cm wide bands which each are set up like a flag on a stick and cut so that it extends 25 cm from the pole. It is then tested by an artificial wind of about 100 km per hr. (II) and (III) delaminated within a few minutes, while (I) withstood delamination, except at the edges, for the 2 hour period

When sample (I) is looked at, no matter from which side, it gives the impression that the coloured strands are a few mm distant from the striations.

The cross-section of (I) is examined in microscope, and fig. 2a is a reproduction of a digetal microphoto of this section. ~~As it appears~~ It exhibits a regular pattern of thickness variations and a corresponding waving, however to a very small extent. In example 1 of the above mentioned European patent this structure, which there is called "U-Rib Structure" is produced much more pronouncedly, see fig. 1 of that patent. The "U-Rib Structure" is defined in claim 9 of this present Patent Application. In the present example the intent has been to form "U-Rib Structure" much less pronouncedly by modified process conditions, namely the bigger angle of cutting, the milder conditions of cooling after the longitudinal stretching, and a slightly higher transverse tension during the annealing.

As it appears from the microphoto the thickness of the laminate varies by about +/-19% in a generally regular pattern, while the angle between the surface referred to as A and the average plane of the surface varies by about +/-3 degrees within a division, also in a generally regular pattern. Due to the influence on the reflection of light, these relatively small variations of angle give the impression of a promounced striation.

There follows extracts from copending British patent Application No. 0205021.9, in which reference

A new cross-laminate comprises mutually bonded polymer films of which at least two neighbour films (A and B) are uniaxially oriented, or unbalanced biaxially oriented, A and B each being coextruded films and each containing a layer consisting of a polymer material selected fro high tensile strength (hereinafter the main layer) and a surface layer (hereinafter the first bonding layer), consisting of a polymer material different from the material from which the main layer is made and selected to produce or enhance the bonding between A and B, whereby the main direction of orientation in A crosses the main direction of orientation in B, and in which the bonding between A and B is strong in spots or lines, while there is no bonding or only a weak bonding between A and B over the rest of the contacting surfaces, <u>characterised in that</u> each of the said coextruded first bonding layers is a discontinuous layer consisting of an array of coextruded thin strands, the array of strands on A crossing the array of strands on B, whereby A and B have become strongly bonded to each other in each spot where a strand on A intersects with a strand on B, while A and B are weaker bonded or not bonded over the parts of their contacting surfaces, which are devoid of any first bonding layer.

A very important advantage of the method and the resulting crosslaminate is that the pattern of lamination comprises not only two, but three elements, provided the coextrusion apparatus comprises the means for extruding the mentioned "second bonding layer" (even when the coextrusion apparatus with said means are not always used to extrude this layer).

These three elements in the pattern of lamination are:

a) each spot where two strands of "first bonding layers" intersect each other,

b) each little area in which both contacting surfaceparts are devoid of any "first bonding layer", and

c) the areas in which there is "first bonding layer" on one of the two contacting surfaces and no "first bonding layer" on the other one.

a) and c) together form the net pattern.

By adapting the bonding strength a), b) and c) differently for different uses, but using the same machinery, this bonding system can be very helpful for "taylormaking" of the crosslaminate.

Thus, as an example, there are certain tarpaulinlike uses where the gauge should be brought down as much as possible for cost reasons, tear propagation strength and ultimate tensile strength is of primary importance, yield tension is of relatively low importance, the estetics unimportant, but the resistance to delamination must be very high due to flapping in the wind. In that case a strong bond/no bond pattern is preferable, and the coextrusion of "second

bonding layer" is omitted. (The main component may be applied not only from its own extruder and through its own channel system, but also from the extruder and through the channel system which otherwise is used for the "second bonding layer"). The bonding is established as a strong welding in the spots (a) where the strands intersect each other.

In other cases there can be a need to establish a strong bonding not only in the spots (a) but also in the areas (c), while there should be some bonding, but a pronouncedly weak bonding in the areas (b).

This can also be achieved by a suitable choise of polymer materials for the "first" and "second" bonding layers (in this case the "second bonding layer" must of course be applied). The combination of strong bonding in a net pattern, and some but weak bonding over the rest of the area is a very interesting pattern of lamination, usually better than strong spotwelding combined with weak bonding over the rest. In the last mentioned case an accidentially started delamination will generally propagate over a wide area if the crosslaminate is repeatedly flexed, e.g. when it is flapping in the wind. The films will still be held together where they are spotwelded, but the rest will become unbonded and thereby loose the estetics and to some extent the yield strength and creep resistance.

Unlike this, a weak bonding surrounded by strong bonding in net pattern, will not be allowed to propagate an accidential delamination in similar manner.

However, it should be mentioned that there also exist

applications in which the best combination is:

(a): strong welding

(b): weak bonding

(c): also weak bonding, but stronger than (b).

Preferably each of the two films A and B should mainly consist of polyethylene or polypropylene, e.g. the main layer can advantageously consist of HDPE or LLDPE or a blend of the two, the second bonding layer mainly of LLDPE but with admixture of 5-25% of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-80 degr. C, while the strands mainly can consist of a copolymer of ethylene having a melting point or a melting range within the temperature interval 50-100 degr. C or a blend of such copolymer and LLDPE containing at least 25% of the said copolymer.

The distance from middle to middle of neighbour strands in each array should normally be between 2 mm and 8 cm, preferably no higher than 6 cm, and more preferably no higher than 2 cm.

The bonding strength in the spots (a) as measured by peeling should normally be at least 40 gram per cm and the bonding strength in areas (b) similarly determined at the highest 75%, but preferably no more than 50% of the bonding strength in (a).

Unlike the crosslaminate made with counterrotating dieparts and comprising criss-crossing arrays of ribs (mentioned above in the section re. known art) the thickness increase in each of the films A and B at the locations where the strands are coextruded, should normally amount to at the highest 30% seen relative to the immediate surrounding, preferably at the highest 20% and still more preferably no more than 10%.

The coextrusion of at least one of the films A or B is preferably carried out by means of a circular coextrusion die, to form and draw-down a tubular film. Hereby the draw-down is adapted to produce a significant uniaxial or unbalanced biaxial meltorientation with the main direction of orientation and the direction of the array of strands either extending along the longitudinal direction of the film or, by means of a relative rotation between the exit of the die and means to take up the film after the extrusion is brought to extend helically along the tubular film.

Subsequently the film is cut open under an angle to the main direction of orientation and to the direction of the array. However, it is also within the scope of the invention to extrude both films A and B from a flat die and cross-web the films under use of a hot press, preferably after longitudinal cold-stretching of both.

The crosslaminate of the present invention is not necessarely limited to the two films A and B, but can comprise 3 or more layers. Thus as an advantageous construction, it may comprise two pairs of array-bonded

films A and B, especially in the arrangement A-B-A in which B has on both of its surfaces an array of strands, i.e. a "first bonding layer" and preferably also a "second bonding layer".

In another suitable arrangement comprising more than two films A and B there is additionally applied at least one more film in the lamination. Said film is also produced by coextrusion and is thereby provided with a surface layer of a composition adapted to control its bonding in the laminate. This composition and the lamination conditions are choosen such that the strength of this bonding becomes higher than the bonding strength between A and B at the locations which are devoid of the coextruded strands. Thus a delamaination of the additional film is counteracted.

The surfaces of the laminate should preferably each consist of a layer adapted to improve the heatseling properties of the laminate and/or increase its frictional properties. Such layers are coextruded in the films used as outer films in the laminate.

Normally the molecular orientation in each film A and B, which may be uniaxial or unbalanced biaxial orientation, should not be limited to that achieved in connection with the extrusion. There may be carried out a further longitudinal stretching prior to the helical cutting. Alternatively or supplementary, the films may be further oriented by stretching in the longitudinal and/or in the transverse direction following the bringing-together of the films in a sandwich arrangement for lamination. This may take place after the heatbonding of said sandwich

arrangement to a laminate.

In the foregoing it was stated that commonly used machinery for manufacture of crosslaminates can be applied, only with inexpensive additions to existing coextrusion lines. This concerns the coextrusion of the array of strands, the discountinous "first bonding layer". The inventor has found that this can be done by adding special but rather simple and cheap machineparts at the exit of almost any existing design of coextrusion dies. Of course there is also need for one more extruder, but the strands will normally occupy only about 2-5% of each of the extruded films, and therefore this can be a small and inexpensive extruder.

The extrusion die according to the invention is a circular extrusion die comprising a distribution part in which at least a first molten polymer material can be formed into a generally even circular flow, and bodily separate from this an exit part comprising a circular main channel with generally cylindrical or conical walls, which channel may comprise a flat zone, to conduct said molten polymer material towards an exit orifice from which it will leave the die as a tubular film structure. The special features of the invention is that said exit part also comprises a channel system for circumferential extrusion of a circular array of narrow strands of a second molten polymer material, this channel system ending in a circular row of internal orifices in the outward generally cylindrical or conical wall of the main channel.

In a prefered embodiment, the circumferential extrusion

starts at one or a few inlets to the exit part and comprises for equal dividing a labyrintine channel system starting at each inlet, each such system comprising at least three channel-branchings.

The term "labyrintine dividing" was introduced in US Patent 4,403,934 and means a dividing of flows in which one divides into two branches of equal length, each of these again into two of equal length etc., all branches mainly being circular and parallel to each other. This is shown in fig. 15

In order to make a particular short distance between the internal orifices, the channels of the labyrintine system or systems may terminate in a common circular channel having a wall common with a part of the generally cylindrical or conical wall of the main channel. The circular row of internal orifices is located in said wallpart.

This coextrusion die has been conceived with a view to the manufacture of the crosslaminate as means to achieve strong bond/weak bond or strong bond/no bond lamination patterns, and for this purpose there is normally a need for a continous "second bonding layer". Therefore, there is preferably in addition to the means for coextruding the said first and second molten polymer materials, provided means for coextruding a circular flow of a third molten polymer material on the side of the first material which is opposite the second material. Channel arrangements for joining the flows of first and third materials are provided either in the said distribution part, or in a part between the latter and the bodily separate exit part.

In the die, the circumference of the inward wall at the exit is preferably at least 20 cm, and the distance from middle to middle of neighbour orifices in the circular row is adapted to produce, after the magnification or reduction which will happen if the walls of the main channel are generally conical, a distance from middle to middle of neighbours of the strands which is at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm.

The die can also advantageously be applied in the production of polymer film other than crosslaminates, e.g. film with a decorative pattern of coloured stripes. Besides the above mentioned economical advantage, namely that existing die designs can be used with addition of cheap dieparts and a small extruder, it is also an advantage that the route of flow from extruder to die exit becomes shortest possible when the inlet to the die and the entire distribution system is near the exit from the die, whereby degradation of the polymer best possible can be avoided.

For the sake of completeness it should be added that the array of strands in the coextruded films A and B for the described crosslaminate, of course also can be formed from a flow which passes through the entire distribution part of a coextrusion die parallel with the other flows, but then there can be a risk of degradation since these strands as mentioned above usually only will constitute about 2-5% of each film.

For each film the main direction of orientation is shown almost but not quite parallel with the array of strands in the film. If the tubular film, when leaving the extrusion die, has been drawn-down straight, and if it has not been stretched after the helical cutting, the main direction of orientation will be exactly parallel with the array, but if the tubular film has been screwed during the drawn-down, to produce a helical melt-orientation, or if it has been uniaxially or biaxially oriented after the helical cutting, as in the above-mentioned US 4,039,364 (Rasmussen), the main direction of orientation will not be quite parallel with the array.

In the spots (a) where one array crosses the other one, there is established a strong spot welding. It should normally be so strong that the laminate will rupture around these spots if delamination is tried.

If there is not coextruded a continuous bonding layer (in the claims called ("the second bonding layer") between the strands and the main layer, there will only be bonding in the spots (a), but as mentioned this will be a very strong bonding. As it has been explained in the general description, this simple bonding system is preferable in some cases. However, the coextrusion die should always comprise a channel system for "the second bonding layer" so that is use is not limited to the said cases.

Most normally there should be coextruded a "second bonding layer" in each film between "main layer" and the array of strands. In the areas marked (b) thee is direct adhesive connection between the "second bonding layers" in the two films, and by the choice of material composition and laminating temperature there is established a predetermined, well controlled weak bonding here.

In the areas (c) the stands on one film are adhesively connected with "second bonding layer" on the other film. Thus the bonding strength in areas (c) gets a value somewhere between those in (a) and those in (b). There is a wide choice between these three values, and the adhesives system can so to say be "taylor made" for the intended use of the cross-laminate.

With relatively thin film it is possible to make the bonding in the (c) areas so strong that it cannot be eliminated without rupture of the material, even when the bonding in the (b) areas is made particularly weak. Hereby these areas of weak bonding are fully "encased" in areas of strong bonding. The advantages of this for certain uses are explained in the general description. In other cases, it can be preferable, still while making the bonding in the (b) areas very weak, to give the bonding in the (c) areas a suitable "in-between" value which allows delamination during tear propagation, but exerts so high resistance against this delamination, that tear energy is absorbed and rupture around the (a) spots is prevented.

Example (of GB Application 0205021.9)

The procedure is the same as in US 5,028,289 (Rasmussen) example 3 except for the following:

The coextrusion line is constructed as shown in figs. ~~2~~ 3, ~~3~~ 4, 4a 3a and ~~4~~ 5, and there are coextruded strands consisting of a metallocene (catalysed) copolymer of ethylene and octylene having melting range 50-60°C and melt flow index 1.0.

The layer which in said US patent is called "lamination layer" and here "2nd bonding layer" is a blend of 90% LLDPE and 10% of the low melting copolymer. The LLDPE has density 0.92g.ml and melt flow index 1.0. The "main layer" and the "heat seal layer" (for heat-sealing of the final cross-laminate) are the same as in the said example 3. The "main layer" forms 75%, the "heat-seal layer" 15%, the "2nd bonding layer" 8%, and the strands 2% of the film.

The angle of cutting is 57°

The temperature for lamination, stretching processes, and final heat treatment are also different, namely:

For pre-heating: 60°C

For the transverse stretching between the special grooved rollers described in the said US patent and the first longitudinal stretching process: 35°C.

For the following transverse and longitudinal stretching processes: 35°C.

For the heat treatment which effects the final bonding: 90°C.

Like in the said example 3 the gauge of the final cross-laminate for identification below called I is about $70gm^{-2}$. This has the bonding pattern shown in fig. 1.

A similar cross-laminate (called II) but without the strands, is manufactured for comparison.

Furthermore there is manufactured a third laminate (called III) similarly to I, but without the strands and with 15% instead of the 10% metallocene copolymer in the "2nd bonding layer".

(I) exhibits the highest tear propagation resistance, (II) almost the same, and (III) a considerably lower tear-propagation resistance under shock-tearing unacceptable for tarpaulins. This property is evaluated by "hand-tearing" at a measured velocity between 5 - 7ms^{-1} by a team of people used to such testing and knowing the requirements of customers. To the knowledge of the inventor there does not exist any standardised test for tear-propagation-resistance which come close to the practical conditions of tearing. (I), (II) and (III) are also cut into 8cm wide bands which each are set up like a flag on a stick of diameter 25mm and cut so that it extends 25cm from the pole. It is then tested by an artificial wind of about 100 km per hr. (II) and (III) delaminated within a few minutes, while (I) withstood delamination, except at the edges, for the 2 hour period which the test lasted.

CLAIMS

1. A crosslaminate comprising mutually bonded polymer films of which at least two neighbour films A and B each being coextruded films are uniaxially oriented or unbalanced biaxially oriented, whereby the main direction of orientation in A crosses the main direction of orientation in B and each contains a layer consisting of a polymer material selected for high tensile strength (hereinafter the main layer) and on each mainlayer on its side which faces the neighbour film A or B at least a first surface layer, characterised in that said first surface layer on each of the films A and B is a discontinuous layer consisting of an array of coextruded thin strands, the strands on A being arranged to cross the strands in B, and the strands consisting of a material which is selected to modify the properties in the surface of the respective film, this modification concerning either the optical appearance of the laminate or the bonding between A and B.

2. A crosslaminate according to claim 1, characterised in that the width and/or thickness ~~thickness~~ of the strands in each of said films A and B is selected so as to occupy amounts at the highest to 30%, preferably at the highest 20% and still more preferably no more than 10% of the surface areas of the respective film and/or the thickness of the respective film.

3. A crosslaminate according to claims 1 or 2 characterised in that the distance from middle to middle of neighbour strands in each array is between 2 mm and 80 mm, preferably no higher than 40 mm, and more preferably no

4. A crosslaminate according to any of the preceeding claims, characterised by a second surface layer on the mainlayer of at least one of the neighbour films A and B on the side of the mainlayer which faces the other one of the said neighbour films, this second surface layer being continuous and either located between the mainlayer and the first surface layer or on top of the first surface layer, and preferably selected for control of the bonding between A and B.

5. A crosslaminate according to any of the preceeding claims, characterised in that it comprises on one or each of the outer films of the laminate, a surface layer which also is surface layer of the laminate and is adapted to enhance heatsealing of the laminate and/or increase its frictional properties.

6. A crosslaminate according to any of the preceeding claims, characterised in that the main layer at least of each of the said two films A and B mainly consists of polyethylene or polypropylene.

7. A crosslaminate according to any of the preceeding claims characterised by the following further features:

a) it has an general thickness of at the highest about 0,3 mm,

b) A forms one surface of the laminate,

c) the laminate surface at least on the A-side

exhibits a visible pattern of striations along one direction

thickness variations in A, the divisions in said pattern being at the highest about 3 mm,

d) the thin strands are coloured, and the rest of the film A is sufficiently transparent to show the coloured strands when the laminate is observed from the A-side, whereby the depth of the corrugations is sufficient to make the strands appear as being at least about 0,5 mm distant from the striations.

8. A crosslaminate according to claim 7 , characterised in that the colour of the strands is formed by a pigment which supplies a metallic lustre or a iridescent effect.

9. A crosslaminate according to claim 7 or 8, characterised in that viewed in a cross-section perpendicular to the striations, the laminate exhibits a generally regular arrangement of ribs which are thicker than the average thickness of the laminate and have a generally concave and a generally convex surface to form a bending of the rib transverse of its longitudinal direction and in that the material in or adjacent to the boundaries of the ribs in the tensionless state of the material are bent in the opposite direction to the rib to give the material between the two adjacent ribs a generally straightened-out shape.

10. A crosslaminate according to any of the preceeding claims and in which strong bonding is established where the strands intersect, while by means of a second surface layer between the strand formed first surface layer and the mainlayer in each of the films A and B, a weak bonding or a

blocking is established in the areas which are devoid of strand material, characterised in that said weak bonding or blocking is established by means of an polymer addition in the 2nd surface layer of a polymer type as used as addition in cling film.

11. A method of manufacturing a crosslaminate comprising mutually bonded polymer films of which at least two neighbour films (A and B) each are formed by coextruding in a flat or circular die a main layer of a polymer material which is selected for high tensile strength ~~(hereinafter the main layer)~~ and a first surface layer ~~(hereinafter the first surface layer)~~ from a polymer ~~different~~ material, and in which A and B each is supplied with a uniaxial or unbalanced biaxial molecular orientation at any stage after the joining of the different materials in the coextrusion die and before the lamination, and prior to the lamination A and B are arranged in such a way that the main direction of orientation in A will cross the main direction of orientation in B, and during the lamination the bonding between A and B is established at least in part through heat, characterised in that in the coextrusion each of the said first surface layers is made discontinuous in the transverse direction, whereby it consists ~~consisting~~ of an array of strands, and in the lamination the array of strands on A are arranged to cross the array of strands on B, and further characterised in that the material from which the strands are extruded is selected to modify the properties in the surface of the respective film, this modification concerning either the optical appearance of the laminate or

12. A method according to claim 11, in which the coextrusion of at least one of the films A or B is carried out by means of a circular coextrusion die, to form and draw-down a tubular film, characterised in that the draw-down is adpated to produce a significant uniaxial or unbalanced biaxial meltorientation with the main direction of orientation and the direction of the array of strands either extending along the longitudinal direction of the film or, by means of a relative rotation between the exit of the die and means to take up the film after the extrusion, the main direction of orientation is made to extend helically along the tubular film, and subsequently the film is cut open under an angle to the main direction of orientation and to the direction of the array.

13. A method according to claim 12, characterised in that the distance from the middle to middle of neighbour strands at the exit from the extruder is at the highest 8 cm, preferably no higher than 4 cm and more preferably no higher than 2 cm, and the circumference of the tube at this exit is at least 20 cm.

14. A method according to any of the claims 11-13, characterised in that following the bringing-together of the films in a sandwich arrangement for lamination, before, ~~or~~ or simultaneously with after the bonding of said sandwich arrangement to a laminate by heat, the films are further oriented by stretching in the longitudinal and/or in the transverse direction.

15. A method according to any of the claims 11-14, characterised in that in the lamination process the strands in A are directly sealed to the strands in B.

16. A method according to any of the claims 11-14, characterised in that the lamination process is extrusion lamination whereby the bonding is established by means of a separately extruded layer.

17. A method according to any of the claims 11-16, characterised by the following further features:
a) the thicknesses of the films used to make the laminate and the stretch ratios are adapted to give the final laminate a general thickness of at the highest about 0,3 mm,
b) A is applied as one surface of the laminate,
c) the laminate surface at least on the A-side is supplied by means of embossment with a visible pattern of striations along one direction constitued by surface corrugations with corresponding thickness variations in A, the divisions in said pattern being at the highest about 3 mm,
d) the material for the thin strands is coloured, and the rest of the film A is maintained sufficiently transparent to show the coloured strands when the laminate is observed from the A-side, whereby the depth of the corrugations is made sufficiently deep to give the strands the appearance of being at least about 0,5 mm distant from the striations.

18. A method according to claim 17, characterised in that

laminate, when they have been brought together for lamination, before or after establishment of the bonding, through one or more pairs of mutually intermeshing grooved rollers, by which the embossment process also becomes a stretching process.

19. A crosslaminate according to any of the claims 1-10, characterised in that said first surface layer on each of the films A and B occupies at the highest 15%, preferably at the highest 10%, and more preferably at the highest 5% of the volume of the respective film A or B.

20. A crosslaminate according to any of the claims 1-10 or 19, characterised in that the average melting point of the polymers which constitute the strandformed first surface layer, is at least about 10^{0} C, preferably at least 15^{0}C, and more preferably at least about 20^{0}C lower than the average melting point of the polymers which constitute the main layer.

21. A method according to any of the claims 11-18, characterised in that said first surface layer on each of the films A and B occupies at the highest 15%, preferably at the highest 10%, and more preferably at the highest 5% of the volume of the respective film A or B.

22. A method according to any of the claims 11-18 or 21, characterised in that the average melting point of the polymers which constitute the strandformed first surface

and more preferably at least about 20^{0}C lower than the average melting point of the polymers which constitute the main layer.

23. Any combination of apparatus which is suitable for carrying out the method according to any of the claims 11-18, 21 or 22.

Fig. 1

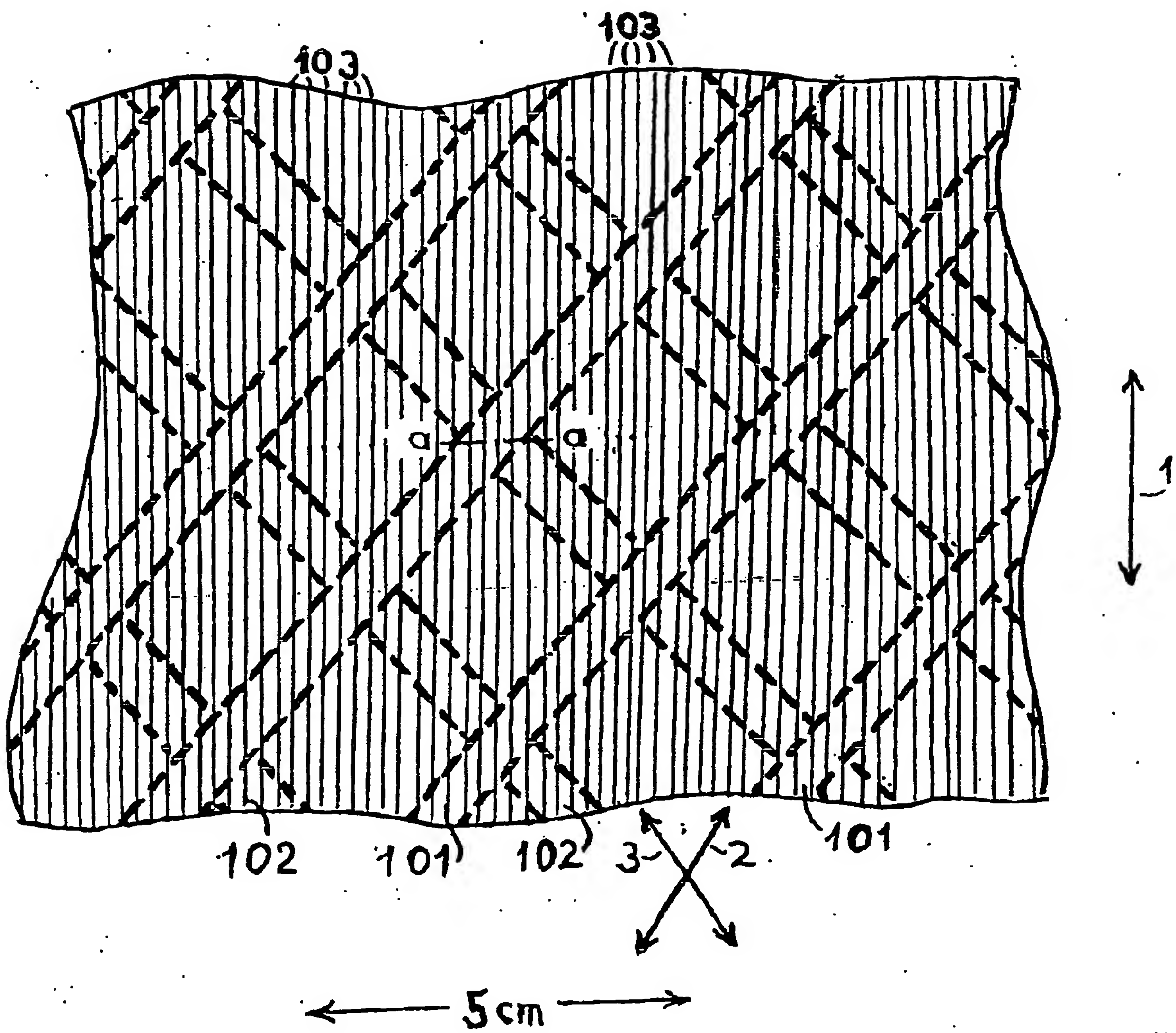

Fig. 2

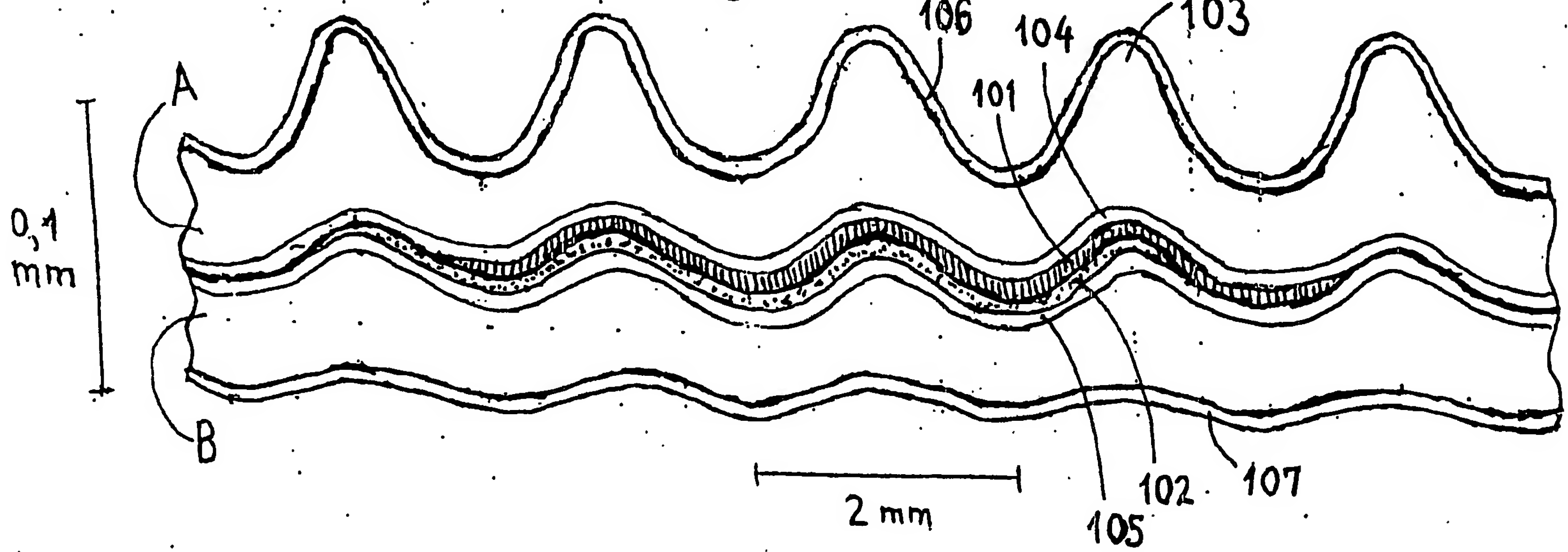

Fig. 2a

Fig. 3

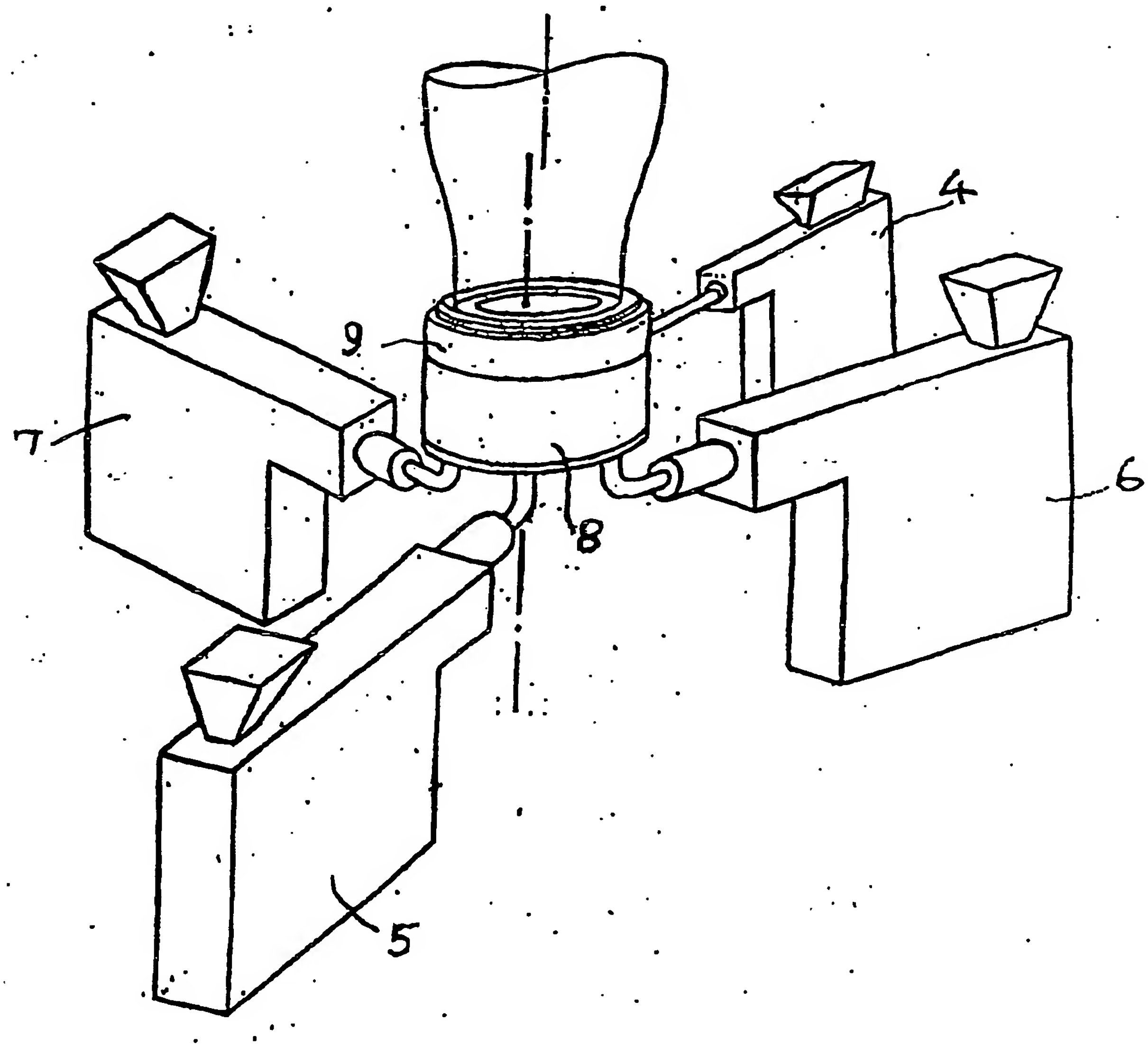

Fig. 4

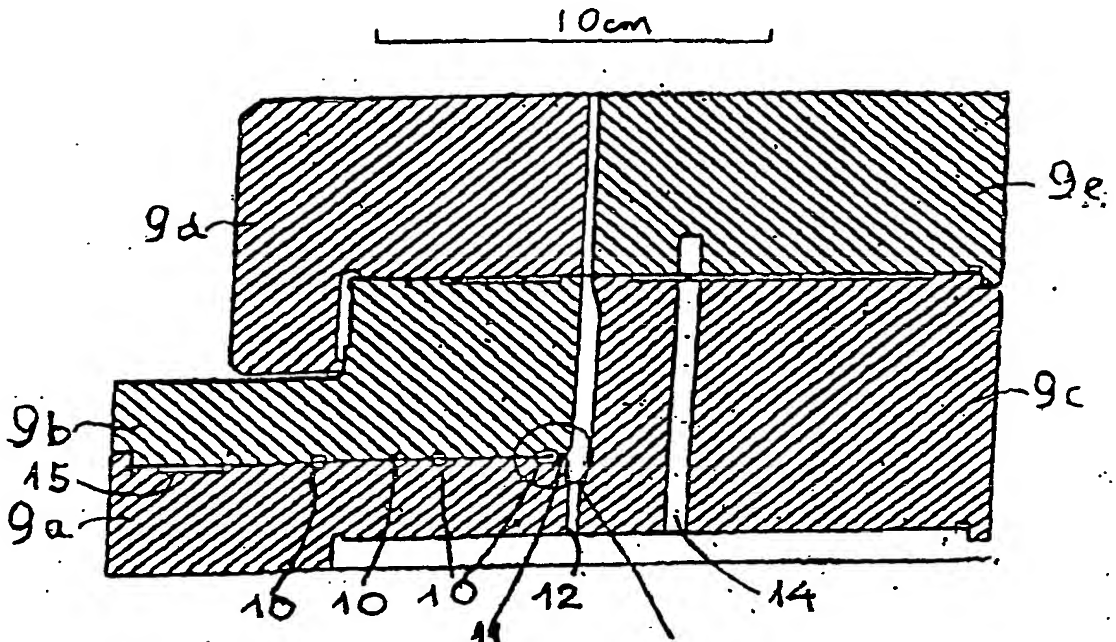

Fig. 3a

Fig. 4a

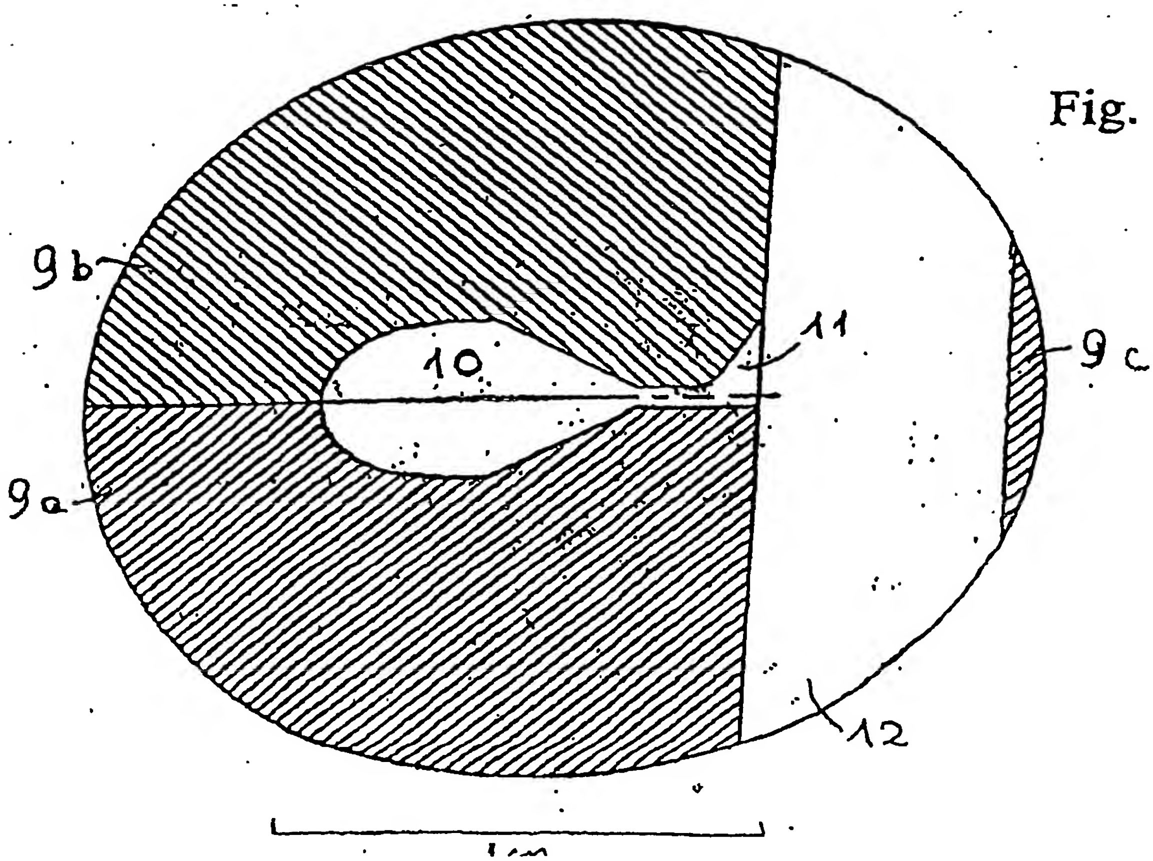

Fig. 5